

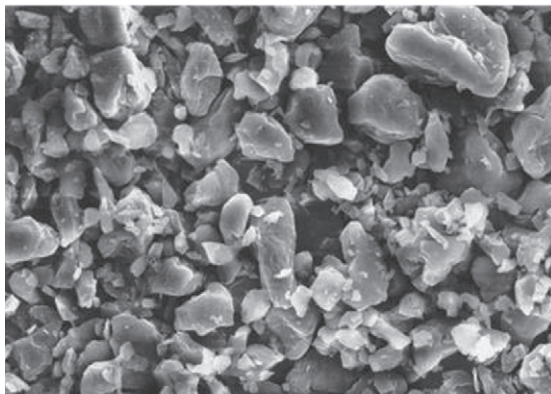
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Evaluation, Airborne Release Fractions, and

Control of Beryllium Hazards at

Los Alamos National Laboratory



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Proposed beryllium metal bounding airborne release fractions (ARFs)/rates (ARRs) and respirable fractions (RFs) for DOE facility accidents analyses

By Jofu Mishima, Terry L. Foppe, J.C. Laul

Beryllium metal has special properties for nuclear applications and is used widely within the Department of Energy (DOE) complex. Beryllium is toxic and has the potential to cause health effects (sensitization and chronic beryllium disease) to workers. Due to the type of processing involving large quantities of beryllium metal and powders and potential for significant airborne release under accident conditions, a comprehensive hazard analysis is required by DOE in a safety analysis document that evaluates potential accidents that can cause beryllium releases to the workplace and public or to the environment. The goal of the safety analysis is to identify and evaluate hazards so that appropriate controls or safeguards can be specified to protect the workers and public from potential accidents. A hazard assessment is also required to comply with 10 CFR Part 850, *Chronic Beryllium Disease Prevention Program* (1999) to protect the workers.

DOE safety analyses include a quantitative estimate of releases and downwind concentrations for purposes of hazard classification and determination of the need for additional hazards analysis, accident analysis, and risk assessments. There are no complex-wide accepted Airborne Release Fractions (ARFs) and Respirable Fractions (RFs) that can be used to evaluate the potential downwind consequences of releases of beryllium under various accident conditions (e.g., spill, fire, stress, explosion, etc.). The purpose is to recommend DOE complex-wide ARFs and RFs that can be used to evaluate the potential downwind consequences of accidents.

This paper proposes various values for ARFs and RFs that are based on review of the published information and data on the oxidation and ignition behavior of beryllium metal encased in a thin coat of non-porous, adherent oxide ("blue oxide"); the typical form of this material found in DOE facilities. Information is also presented as a function of the morphology of the metal (i.e., large, coherent, pieces; turnings/swarfs; chips/powder; and dust) because the surface to volume ratio is an important factor in evaluating both the amount of beryllium released and the potential for ignition of the metal that may increase the amount of release significantly.

Guidance is also provided for the use of the DOE-HDBK-3010-94, *Airborne Release Fractions/Rates and Respirable Fraction for Nonreactor Nuclear Facilities*, values for oxide releases that result in a more liberal, allowable limiting airborne concentration for determining consequences and hazard classification of the facility. Detailed explanations of the proposed ARF/RF values for the various beryllium metal forms (large coherent pieces, chips/powder, turning/swarfs from processing, and dust) and other aspects of beryllium are provided in a report by Mishima et al. (April 2005). A short version of this report and a summary of the ARF/RF values are presented here.

INTRODUCTION

Beryllium (Be) metal has special properties for use with nuclear materials (high tensile strength, light-weight, and absorbs/reflects neutrons).¹ These properties influence the handling and processing (metal shapes most commonly found in the Department of Energy (DOE) complex such as formed by compaction of metal chips, furnace casting and machining) of the material.

Beryllium is a toxic material and an inhalation hazard causing sensitization and chronic beryllium disease (CBD) to receptors.² Beryllium health effects, exposure limits and regulatory requirements on sensitization and CBD for workers and the public are discussed by Wambach and Laul.³ Releases of hazardous chemicals from DOE facilities have the potential to cause significant consequences to receptors and must be assessed with a rigor and completeness equal to the analyses for the releases of radiological materials. The beryllium hazard evaluation and consequence analysis during potential accident aspects are discussed by Laul and Norman.⁴ The primary concern is the airborne release of the oxide resulting from abnormal, accident situations. The beryllium metal (metal core encased in "blue oxide" film) found in DOE facilities are typically in various forms of metal (large coherent pieces, chips/powder, turning/swarfs from processing, and dust) and its compounds are covered in this paper. The concentration guides

for beryllium oxide is applicable in the circumstances where the airborne release is due to heated or ignited metal but bear in mind that under some situations such as involving a large quantity of beryllium, conversion to the oxide may be incomplete and some metal particles may be released. Information is provided for other beryllium compounds to provide a comprehensive view of a specific topic but the compounds are not discussed with the intent to define ARFs or RFs fractions for those materials.

The term "beryllium metal" is really a misnomer; all "metal" except during the actual machining operations, in DOE facilities is pacified by a protective, non-porous, tightly adherent, very thin, beryllium oxide ("blue" oxide) film. The term "pacified" is used to mean to severely attenuate the passage of oxygen to the metal surface and, thus, prevent or severely reduce the oxidation of the metal under this protective layer. Also note that the term "dust" in this report is intended to denote a relatively fresh layer of the kind found on metallic surfaces in processing facilities. It is not intended to represent bulk powders or material entrained in waste handling streams. The former should be considered powders, while latter should be characterized as large coherent pieces, powders or chips as is appropriate. Furthermore, note the inherent difficulties in characterizing the particle size of extraneous beryllium metal that is necessary to consider material as "dust" (geometric diameter [d_G] <20 micrometers [μm]).

PURPOSE

The primary concern is, therefore, the airborne release and suspension of beryllium and its' oxide. Even for dust that is of the size range that may be suspended, the individual particles are composed of oxide over a metal core that constitutes a larger fraction of the mass. Published data on the physical and chemical properties cited, below, has some attached degree of uncertainty due to the possibility the measurements are altered by the presence of the oxide film and that the protective oxide layer may continue to grow

with time. These facts may also provide an explanation of the varying responses cited for the behavior of beryllium forms.

The behavior of the beryllium metal cited in the references also has some degree of uncertainty attached because the experimental parameters measured may not completely define the controlling parameters for that behavior. The oxidation and ignition of beryllium metal is a complex phenomenon that is reflected in the variation in reported values (even for characteristic such a density, melting point, boiling point, etc. that may include varying quantities of oxide) and the anomalous ignition behavior due to the inadvertent formation of various Be compounds that have not been thoroughly studied chemically, such as organo-metallic compounds, nitrides, carbides, etc., that may be incidentally formed during handling and production.

Thus, the physical (morphology, size, surface to volume ratio, prior handling), chemical characteristics, and the environment, all have an effect on the oxidation and ignition behavior of beryllium metal. Knowledge of these aspects provides a better basis for increased understanding of the potential behavior under accident conditions postulated.

This paper provides information on:

- the literature describing physical and chemical properties of beryllium metal and beryllium oxide;
- experimentally measured responses of encased-beryllium metal to various conditions (principally heat and morphology);
- the technical bases for the ARFs and RFs cited in document; and,
- guidance on the applicability of DOE-HDBK-3010-94⁵ ARF and RF values for beryllium oxide behavior under the stated conditions.

The descriptions and information on accidents that have resulted in the release of airborne beryllium are discussed in Mishima et al.⁶ to illuminate the potential conditions and situations that may lead to ignition and release of encased-beryllium metal forms, however, are not presented in this paper.

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BERYLLIUM CHARACTERISTICS

Although discovered in 1798, beryllium (Be) metal was not widely used until the past few decades. Beryllium is used for alloying (contributes hardness, strength, high electrical and thermal conductivity, and resistance to wear, corrosion and metal fatigue) and its oxide, BeO, used as an electrical and thermal insulator. For beryllium metal, its thermal conductivity is 201 J/m s with an electrical conductivity of 250 mohms/cm.¹ The element forms many salts (e.g., fluorides, chlorides, sulfates, nitrides, carbides, etc.). Its atomic mass is 9.0122 and has an isotope ⁷Be with a half life ($t_{1/2}$) of 53.3 days (d). Beryllium is classified as an “Alkaline Earth Metal”. Its crystal structure is hexagonal. Except for magnesium, it is one of the lightest metals. Beryllium is not an “ideal” metal – it is expensive, brittle and toxic.

Allowable Air Concentrations

The various air concentration limits for beryllium and its compounds are taken from the U.S. Environmental Protection Agency (EPA)⁷ and are shown in Table 1. The concentration limits of concern for the purposes of this paper are the Emergency Response Planning Guidelines (ERPGs) and the Temporary Emergency Exposure Limits (TEELs).

The various limiting airborne values indicated are applicable to beryllium that is released at room temperature. The limiting airborne concentration for beryllium oxide are applicable to beryllium released under thermal stress and are a factor of 25–100 greater than the airborne concentration limits for beryllium metal.

Table 1. ERPGs/TEELs Values for Beryllium and its Compounds

Compound	ERPG-1/ TEEL-1 (mg/m ³)	ERPG-2/ TEEL-2 (mg/m ³)	ERPG-3/ TEEL-3 (mg/m ³)
Beryllium metal, Be	0.005	0.025	0.1
Beryllium hydride, BeH ₂	–	–	–
Beryllium hydroxide, Be(OH) ₂	0.025	0.25	20
Beryllium oxide, BeO	0.0125	1.25	10

Table 2. Cited Physical Properties of Beryllium Metal

Property	Cited Value
Atomic diameter (Å)	2.221
Formula weight	9.012; 9.0122; 9.02
Density, ρ (g/cm ³)	1.85; 1.8477; 1.816
Melting point (°C)	1,278; 1,287; 1,284; 1,283
Boiling point (°C)	2,467; 2,950; 2,767; 2,970
Vapor pressure (atm.)	$\log P = 6.186 + 1.454 \times 10^{-4}T - 16,700/T$
Latent heat of fusion (cal/g)	250/275
Thermal neutron cross-sections (barns)	0.0090 \pm 0.0005
Coefficient of thermal expansion	(See Table II in ref. 14),
Thermal conductivity	(See Figure 4 in ref. 14)
Specific heat	(See Figure 1 in ref. 14)
Enthalpy	(See Table III in ref. 14), 0.62 ^{400–500 °C} ,
Entropy (solid), $S_{298.1}$ (cal/mol K)	2.28 \pm 0.028
Entropy (liquid), $S_{298.1}$ (cal/mol K)	32.56 \pm 0.03
Emissivity coefficient	0.81
Reflectivity (white light)	50/55%
Velocity of sound (m/s)	12,600

Physical Characteristics

Beryllium is a very hard, silver-gray metal. Its physical characteristics are presented in Table 2. There are some variations in values as noted by the various authors.^{1,8–12,14}

Chemical Characteristics

Composition: Information on the composition of Be and other associated elements and on particle size fractions is reported in Lewis and Karlovitz¹³ and shown below:

- Be – 97.6–92.0%;
- BeO – 1.20–8.1%;
- C – 0.08–0.33%;
- Al – 0.05–0.19%;
- Fe – 0.10–0.17%;
- Mg – 0.01–0.15%; and,
- Si – 0.04–0.12%.

Corrosion: “The corrosion resistance of beryllium in air at room temperature is analogous to the excellent corrosion resistance of aluminum under the same conditions”.¹⁴ The volume of the oxide is greater than the volume of the metal consumed. At room temperature, the “blue” oxide film formed is nearly non-porous and thin (nominal thickness is 0.01- μ m and reaches a maximum thickness in 2 hours [hr]). No severe corrosion was observed at <600 °C in air. Hydrated beryllium oxide (BeO· x H₂O) formed on extruded, machined rods exposed for 6 months to ambient air during the spring and summer at Oak Ridge, TN (including days at high relative humidity). They observed blisters and inclusions of corrosion products from the slow oxidation of beryllium carbide inclusions.

Activation energy

- Be (solid) + (1/2)O₂ (gas) \Rightarrow BeO (solid) + 136 kcal.
- BeO and Be₃N₂ are stable between 0 and 1000 °C. The reaction between sintered Be and O₂ is slow between 350 and 950 °C. Activation energy: 50.3–70 \pm 15 kcal/mol.¹⁵
- Activation energy for oxidation 50,300 cal/mol.¹⁵

Explosibility: Studies on the explosion potential for airborne beryllium fines (powders) all report that the material is category S1 – weakly explosive. The individual studies are: Kent,¹⁶ Coleman,¹⁷ Lewis and Karlovitz,¹³ and Jacobson et al.¹⁸

Fire fighting measures: (Material Safety Data Sheet – No. M11, *Beryllium Powders*)

- Ignition point: Layer of powder in size range of 1–5- μm between 540 and 700 °C; <74- μm screen (<200-mesh) will not ignite under like testing.
- Explosive limits: ASTN Standard E1226 test, *Standard Test Method of Pressure Rise for Combustible Dust*, Be powder @ 3000-g/m³ hazard category, S1, “weakly explosive”.
- Extinguishment media: “Do not use water or carbon dioxide” to extinguish beryllium powder fires. As a powder, extinguish by smothering using a Class D fire extinguisher, dry sand, graphite powder, or sodium chloride.
- Unusual fire and explosion hazards: Do not use “water to extinguish fires around operations involving molten metal due to the potential for a steam explosion. In addition, water may dissociate when in contact with burning beryllium powder or chips releasing flammable hydrogen gas which could burn and result in an explosion”.

Behavior under thermal stress: Beryllium metal is not easily fragmented under accident conditions into particles that can be made airborne. The airborne release of beryllium is of a

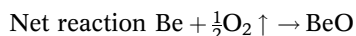
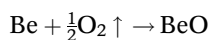
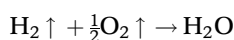
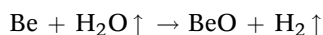
particulate material and the mechanism to convert the metal to this form is by oxidation that is accelerated by thermal stress. Therefore, this discussion focuses on primarily of oxidation and ignition.

Reaction mechanism with water vapor

If water vapor is the only gaseous reactant, the only thermodynamically allowed reaction at the temperature of interest is $\text{Be} + \text{H}_2\text{O}\uparrow \rightarrow \text{BeO} + \text{H}_2\uparrow$. At a temperature of >800 °C a rapid reaction and the surface-reaction rate is proportional to the surface concentration of the water vapor and increases exponentially with temperature.

At sufficiently high temperature [not specified], the reaction is no longer controlled by surface-reaction rate, but by the diffusion of water vapor is through the hydrogen created at the surface of the metal.

With oxygen and water vapor reactant gases:



The reaction results in a decrease of the total moles in the gaseous-phase and, hence involves a bulk flow to the metal surface as the reaction proceeds; reaction is not diffusion-limited. This suggests that the melting-point of oxide or boiling point of metal represent the point at which the transition from surface-reaction to vapor-phase combustion occurs.

BERYLLIUM OXIDE

Physical characteristics: Physical characteristics data, taken from Weast,¹⁰ is shown in Table 3.

Oxidation Process

All metals have a partial pressure of vapors above the surface as a function of temperature. Experimental results have shown that the oxidation of beryllium is controlled by the release of beryllium vapor through the “blue oxide” layer.^{15,19–21,23} The airborne release of significant amounts of the beryllium oxide is primarily concerned with the potential conditions that may result in accelerated oxidation and ignition of the beryllium handled and processed within non-reactor, nuclear facilities. The oxidation process is affected by many parameters that may increase the rate of beryllium metal vapor.

The beryllium oxide-protective film formed at room temperature is composed of very small crystallites and forms a very thin layer around the metal core (d_G is sub-micrometer) that attain a maximum depth of 0.01- μm in 2-hr.¹⁴ The protective layer is a vapor-diffusion barrier but it is unclear whether the depth of the layer continues to grow with time, albeit at a very slow rate, as indicated by the varying oxide concentrations reported.¹⁵

The beryllium-vapor diffuses through protective oxide-film at elevated temperatures and forms an oxide layer with different physical characteristics (a white, “fluffy” therefore a more porous material) than the “blue” oxide-protective layer (1–8%) that is non-porous and tightly adherent, as shown in transmission electron micro-

Table 3. Physical Characteristics

Compound	Formula	Mol Wt.	Crystalline Form	Density (g/cm ³)	Melting Point (°C)	Boiling Point (°C)	Solubility (g/100 cm ³)		
							Cold Water	Hot Water	Other Solvents
Beryllium oxide	BeO	25.01	Wh. Hex	3.01	2530 \pm 30	~3900	0.000002	–	s conc. H ₂ SO ₄
Beryllium oxide hydrate	BeO·H ₂ O	–	Wh. amorp. powder or gel	–	d	–	i	i	s a, alk. (NH ₄) ₂ CO ₃

d: decomposes; i: insoluble.

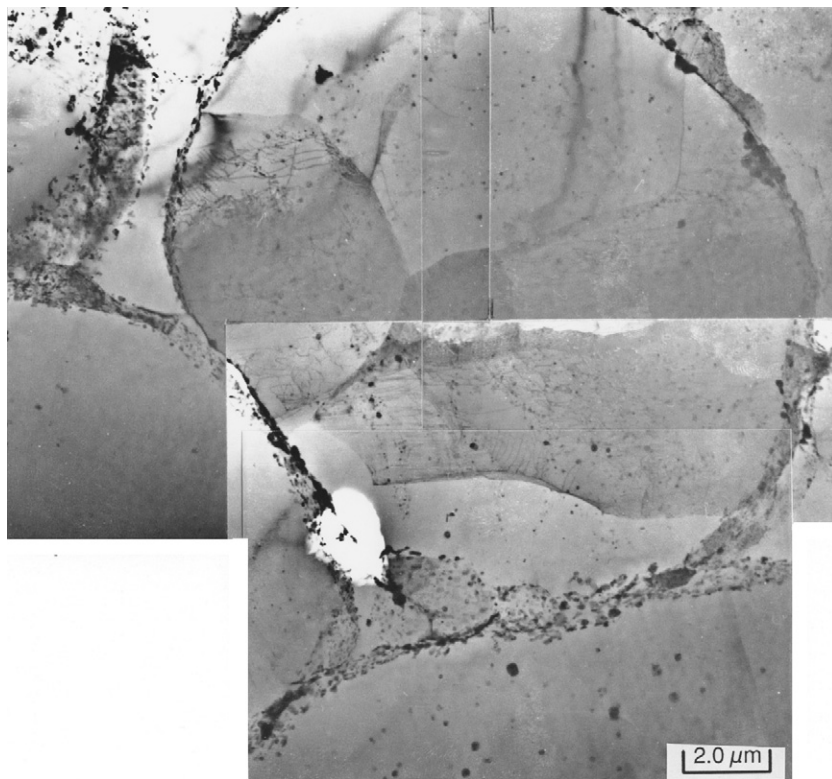


Figure 1. Transmission electron microscope (TEM) of consolidated atomized beryllium powder, showing an individual particle surrounded by a thin continuous ring of oxide (taken from Stephen P. Abeln, 1992).

scope (TEM) in Figure 1. The oxide thickness measured by Auger Microscopy of impact ground three powder particles were 220 Å, 300 Å, and 330 Å with an average at 280 Å.²² Thermal expansion coefficient of Be metal is 11.4 ppm/°C and that of BeO is 9.0 ppm/°C, which causes cracking of the “blue oxide” film on heating. The quantity of the beryllium-vapor released through the oxide layer is a function of the temperature and the surface area. The disturbance (mechanical, such as volume-expansion of metal core by temperature before or at the melting point of the metal) may increase vapor-diffusion and creates a “flaw” in the barrier (protective layer), which leads to oxidation ($2\text{Be} + \text{O}_2 \xrightarrow{\text{heat}} 2\text{BeO}$).

At some temperature; a “hot spot” forms at the “flaw” and rapidly spreads over the entire surface – this is when ignition and self-sustained oxidation occurs. The oxidation-reaction is postulated to occur in the oxide layer and can transfer heat to the metal core. As

the generation rate of vapors increases, oxidation appears to occur further from the oxide layer and morphology of oxide changes – oxide formed by expansion of vapor in ambient environment are small geometric diameter (d_G) platelets/spheres and those formed closer to particle form cylindrical shaped particles. The oxide formed back-diffuses to the surface of the particle of origin and so less than half (cited as 40%) is released from the beryllium metal particle of origin. Due to the decreased bulk density of oxide, the oxide particle remaining is approximately the same size as the original oxide-encased Be metal. Therefore, the beryllium released by thermal stress including during self-sustained oxidation (ignition) is in the form of beryllium oxide. Jordan⁸ suggested significantly lower value of 0.01 for beryllium oxide as a bounding case.

It should also be noted that the ignition temperatures of the various physical configurations cited are, generally

speaking, greater than the anticipated air-temperature generated by fires fueled by solid combustible materials and would require direct flame impingement on the beryllium form for some duration (minutes to hours) to transfer sufficient heat through the refractory oxide layer. Such conditions should not normally be assumed for fires addressed in safety documentation for DOE application (e.g., general room fires, waste handling facility fires, transportation accidents) without a technically sound analysis to support the occurrence of the condition. This means it is not necessary to provide analysis proving that ignition temperatures cannot be reached under any foreseeable conditions to dismiss the possibility; rather, proof must be provided that ignition conditions are a reasonably expected outcome in order to postulate their occurrence. Also bear in mind that the material made airborne is in oxide form that has different ERPG/TEEL values than the metal.

Even the forms such as large coherent pieces that are composed of highly compressed beryllium powder/chips from a manufacturer are pacified by a protective, non-porous, tightly adherent, beryllium oxide (“blue oxide”) film. The protective film is rapidly reestablished over the surfaces (at room temperature) when the film is disrupted.

Beryllium oxidation appears to be a function of the metal temperature, surface to volume ratio and, potentially, age (i.e., thickness of the protective oxide layer that is a refractory material that both hinders the release of beryllium vapors and the transmission of heat to the encased metal core that affects the vapor pressure of the beryllium above the metal). Some gases/vapors appear to enhance the oxidation/ignition such as high water vapor concentration (>20 vol%), hydrogen rich oxygen-hydrogen atmosphere (from the thermal decomposition of water), carbon dioxide, halogens, etc. Many of the experimental studies cited measure the heated air-temperature (furnace-like conditions), not the metal temperature. This is an important factor, because beryllium oxide is an electrical and thermal insulator.

Thermal and Surface to Volume Dependence of Beryllium Metal Oxidation

Both the oxidation (release of Be-vapor through the adherent, non-porous, BeO – “blue” oxide that encases the metal core) and ignition (self-sustained oxidation) are functions of the temperature of the metal core, the thickness of the oxide layer, and the surface area of the metal shape for any local environmental condition imposed. The temperature of the metal core is affected by the transmission of heat energy through the “blue” and white oxide layer and the heat transfer throughout the metal minus heat losses. The thickness of the oxide layer may be affected by the age and prior oxidation, such as a slow rise of temperature that allows growth of oxide layer.

The temperature reported in the experimental data to describe the behavior of the beryllium metal, in most cases, is the heated air-temperature (analogous to placing the material in a furnace), not the metal temperature. This configuration (exposure in a furnace-like volume) is *not* representative of the conditions that beryllium metal is subjected to thermal stress under typical solid, combustible material-fueled fires; unless, the burning material is directly under the metal or transferred by conduction to the surface on which the metal is located. As modeled in codes such as CFAST (Consolidated Fire Smoke Transport), the heated gases rise to the ceiling and the heated layer descends with time. Typically, the gases from these types of fire seldom exceed 600 °C, a level generally observed to cause room flash-over. If the beryllium metal is exposed to the descending layer of hot gases, some time would be required to transmit the heat energy to the metal core; while the oxide layer grows due to the accelerated beryllium vapor release.

A more representative configuration would be the case of radiant heat energy impacting the metal, one side of the metal form is heated by absorption (dependent of the absorption coefficient of the “blue” oxide film) of the radiant heat flux and the energy generated by the oxidation but is lost by convection or conduction on the

Table 4. Comparison of Volume, Weight and Sphere Sizes

d_G Size (cm)	Volume (cm ³) ^a	Weight (g) ^b	Surface (cm ²) ^c
1×10^{-4} (1- μ m)	5.24×10^{-13}	9.69×10^{-13}	3.14×10^{-8}
1×10^{-3} (10- μ m)	5.24×10^{-10}	9.69×10^{-10}	3.14×10^{-6}
1×10^{-2} (0.1-mm)	5.24×10^{-7}	9.69×10^{-7}	3.14×10^{-4}
1×10^{-1} (1-mm)	5.24×10^{-4}	9.69×10^{-4}	3.14×10^{-2}
1	0.524	0.969	3.14
10	5.24×10^2	9.69×10^2	3.14×10^2
100 (1-m)	5.24×10^5	9.69×10^5	3.14×10^4

^a Volume = $4/3[\pi][\text{radius}]^3 = 4.189[r]^3$. ^b Assumes density of Be metal/oxide is 1.85-g/cm³. ^c Surface = $4[\pi][\text{radius}]^2 = [\pi]\text{diameter}^2 = 12.57[\text{radius}]^2$.

surfaces not impacted. The heat absorbed by the metal core is the balance between these two aspects and is a function of the temperature and surfaces.

Gases/vapors and materials that are reported to enhance oxidation/ignition include: >20% water vapor, carbon dioxide, hydrogen rich/water vapor, halogens, water-based solvents, and the formation of organo-metallic compounds. Both carbon dioxide and water vapor are products of the combustion of hydrocarbon fuel.

Since the surface to volume ratio of the metal form plays a dominant role in the airborne release of beryllium, the ratio of various metal forms should be understood. The surface area of the various metal forms (coherent metal, chips/powder, turnings/swarfs, and dust) may vary significantly. The information on the studies to determine the oxidation and ignition temperature of beryllium is segregated into the response by physical shape (i.e., large, coherent pieces; chips/powder; turnings/swarfs; and dust). Table 4 provides the volume and mass associated with spheres of various diameters.

A comparison of surface area as a function of three shapes is shown in Table 5.

Therefore the airborne release for the same weight of encased-beryllium metal should be the greatest for a ribbon of metal (turning/swarf) followed

by a chip. The airborne release from a sphere of encased-beryllium metal should be the minimum value. Since encased-beryllium metal is provided as chips/powder of material from fragmented solids and due to the chemical reactivity of the molten material in air, a spherical shape is not normally found.

OXIDATION – EXPERIMENTAL OBSERVATIONS (LITERATURE REVIEW)

This section provides a review of the experimental data to provide a background on the behavior of oxidation and ignition for the various metal forms of beryllium. Additional information about these experiments and their interpretations is available in the Mishima et al.⁶ report.

The oxidation and ignition of the beryllium metal form are very complex phenomena that may be affected by a variety of local conditions. The oxidation and ignition are affected by the surface to volume ratio of the specific item, the metal core heat input and loss, the thickness of the “blue” oxide-protective layer that may increase with age, etc. The physical configuration of the various sized beryllium metal forms (i.e., large, coherent, pieces; turnings/swarfs, chips/powder, and dust) can vary significantly with the distance separating

Table 5. Aspects of Various Shapes of Beryllium

Form	Dimensions (cm)	Volume (cm ³)	Surface Area (cm ²)	Surface: Volume
Sphere	d_G 1	0.524	3.14	6
Chip/powder	$0.5 \times 0.5 \times 0.5$	0.125	1.5	12
Turning/swarf	$1 \times 5 \times 0.025$	0.125	10.3	82

the individual pieces and would affect the heat balance of the metal core. Furthermore, instances where beryllium metal pieces have spontaneously ignited or have been ignited, cited below, raise uncertainty on the specific conditions that may result in ignition (self-sustained oxidation) and the size classifications for the various metal forms are selected based on best engineering judgment. Therefore, extrapolation of the surface-specific airborne release values for large, coherent pieces suspended in a liquid-petroleum fueled fire to other beryllium metal forms by other categories of fires may have a large degree of uncertainty associated with any value selected.

As discussed in Section 'Purpose', data and information on the behavior of beryllium metal encased in a thin film of adherent oxide is at times contradictory. Blumenthal and Santy²³ concluded that: "*Variation in onset accelerated oxidation and ignition may be due to experimental conditions and size & morphology of specimens ... specimen temperature is the balance of heat from exothermic reaction and losses, depends on particle mass & morphology and experimental configuration ... temperatures cited are typically air temp not specimen.*"

Various models have been postulated to explain the oxidation of beryllium metal encased in a thin film of adherent oxide. General consensus favors the model postulating that the diffusion of Be vapors through the oxide film limits the rate of oxidation. Kuehl²⁴ states that in the vapor-phase model, evaporation is the rate-limiting step. For a heated, single, small ($d_G = 20\text{--}34\text{-}\mu\text{m}$) particles, oxidation appears to take place in the vapor phase.¹⁹

At temperatures above the boiling point (bp) of beryllium, oxide vapors condense as rods up to $10\text{-}\mu\text{m}$ long. Above the melting point (mp) of BeO, vapors condense to droplets with a $d_G \sim 1\text{-}\mu\text{m}$ (the rate of vapor generation appears to eject the vapors a distance sufficient to result in dispersion of the vapors and reaction with air of small amounts of vapor). The oxide particle remaining is approximately the same size as the original encased-metal particle after complete oxidation and contains $\sim 60\%$ of the beryllium mass (the

bulk density of the oxide layer is appreciably less than the metal). There does not appear to be a precise ambient temperature for ignition.²⁵ Blumenthal and Santy²³ proposed the following oxidation and ignition behavior scheme based on experimental data:

- $<1,047^\circ\text{C}$ ($1,320\text{ K}$) – reaction rate slow, oxide film protective, relatively independent of oxygen pressure;
- $>1,047^\circ\text{C}$ – abrupt break-down in protective nature of oxide film after a few minutes, rate of reaction increases as much as $500\times$ (perhaps due to cracking/flaws in oxide coat);
- $1,047\text{--}1,284^\circ\text{C}$ (melting point of beryllium metal) – rate proportional to O_2 pressure;
- $>1,284^\circ\text{C}$ – rapid reaction, oxidation rate proportional to $\sqrt{\text{O}_2}$ concentration;
- @ $1,427^\circ\text{C}$ – $>50\%$ of the beryllium sample is consumed in 15 minutes;
- $>1,047^\circ\text{C}$ + near one atmosphere O_2 pressure, beryllium did not ignite;
- data summarized by Jordan⁸ suggests that the transition from surface-reaction to vapor-phase combustion occurs at temperature $>1,997^\circ\text{C}$... either at the melting point of oxide, $2,527^\circ\text{C}$, or boiling point of metal (reported values $2,397\text{--}2,967^\circ\text{C}$).

The H_2 –air flame system was used to investigate the environment between $\text{H}_2\text{--O}_2$ and CH_4 –air systems. For CH_4 –air flame (max. temperature $2,000^\circ\text{F}$ (1093°C)), a loose, thick, porous oxide layer formed as for $\text{H}_2\text{--O}_2$ flame, only trace quantities of oxide were released and the temperature was similar to value recorded for SNAP (Space Nuclear Auxiliary Power) test discussed later⁸:

- 10-mil foil used to increase metal temperature instead of cylindrical sample.
- Minor hotspots appeared as approached $2,400^\circ\text{F}$ ($1,316^\circ\text{C}$), hotspots 200°F (93°C) to 300°F (149°C) above coldest portion of sample surface ... "*Peak temperatures were, in general, 100°F (38°C) to 300°F (149°C) above the calculated steady-state temperature,*

indicating at least some surface heating due to the reaction. ... The average rate of release of airborne contamination in the experiments with the foil sample was one to two orders-of-magnitude higher than the release rates observed in the experiments with the beryllium cylinder. Because the average peak temperature in the two sets of experiments differed by only 300°F (149°C), it appears that the airborne beryllium release rate is strongly temperature-dependent." Even so, there was little tendency for the crusty oxide coating to become airborne. Oxide consisted of agglomerates up to a few μm in size consisting of small crystalline platelets.

Experimental findings are:

- At the lower temperature, the oxidation rate is approximately independent of oxygen partial pressure in temperature range of $350\text{--}390^\circ\text{C}$.
- Between $1,050$ and $1,284^\circ\text{C}$, the reaction rate is constant and nearly proportional to oxygen partial pressure, but Lindsay and Robinson²⁶ reported that oxidation of chips accelerates rapidly at $1,100^\circ\text{C}$ (near the melting point of beryllium metal) as the oxide layer is disrupted. 2% of beryllium oxidized after 80-minutes @ $1,185^\circ\text{C}$; this agrees with Macek and Semple¹⁹ that accelerated oxidation occurs near the melting point of beryllium.²³ Oxidation of particle accelerates rapidly at 750°C and above.³⁹
- At temperatures $>\text{mp}$ Be metal, the reacted beryllium is encrusted with a thick, white, porous layer of beryllium oxide (metal comes with a thin, dark blue film) made up of agglomerates of small crystalline platelets a few hundred Angstrom [10^{-10} m or $0.0001\text{-}\mu\text{m}$] to several μm thick, and adheres fairly tightly to metal core. "*Reaction products were found a considerable distance from the beryllium sample*"²¹
- Experimental observations are that much of the high-temperature oxidation occurs in the vapor state away from particle surface – oxidation products are airborne at creation, and are fine drops or rods in the

- respirable size range.⁸ Approximately 60% of the oxide from ignition of an isolated particle remains with the particle, as concluded by Macek²⁰: “*The observed sizes of the spheres indicate that a large fraction of the beryllium may end up in the massive central particle – about 60%, if the terminal size of the oxide is the same as that of the original metal particles. This vapor-phase model predicts rates reasonably well.*” The estimate of 60% oxide remaining with original particle may be an under-estimate.⁸ Airborne particles collected from vapor-phase combustion of 4.75-mm × 12.70-mm cylinder in O₂-rich H₂ or O₂ flames exactly like Macek’s (sub-μm spheres and rod-shaped crystals several μm long).²⁷
- Most oxide remained on a 0.5-g Be sample @ 1,500 °C and 1-atm O₂.
 - @ 1,200 °C and 1-atm O₂, the sample does not melt; it is encrusted with oxide, and retains original shape – little, if any, airborne.⁸ @ 1,500 °C and 1-atm O₂, >50% of the specimen oxidized within 15 min [weight/size of specimen not given].²¹
 - At the bp of beryllium, oxidation is rapid and the oxide forms a porous coat (corroborates characteristics of oxide depend on temperature during formation) and airborne release depends on the degree of mechanical disruption by external action.²³
 - At temperatures >mp oxide (~2,547 °C), the oxide layer is severely disrupted and beryllium-vapors condense to d_G 1-μm particles in the vapor state ... some fraction of vapors back-diffuse and condense on remnants of original mass so that the particle remains approximately the same size ... some oxidation in the liquid state may occur.⁸
 - Individual particles in a powder show a localized bright spot on ignition rather than *uniform ignition throughout the mass* corroborating that ignition is from one point rather than uniform ignition.¹⁵
 - Oxidation of sintered and cast metal turnings – Oxidation in the region of 1000 to 1,100 °C slows nearly to a standstill as beryllium oxide film thickens. With occurrence of the metal’s melting at 1,278 °C, however, the protection is lost and the reaction rate accelerates rapidly. Oxidation of sintered and cast metal turning does not become significant much below 1,200 °C. Environmental temperatures in excess of 1,000 °C would appear to be necessary for a burning process to occur. This temperature could conceivably drop to 800 °C for fines [size undefined].²⁶
 - If the reaction occurs above the mp of BeO [2,550 °C], condensation to 1-μm [d_G] drops of beryllium oxide; otherwise, oxide condenses into 10-μm rods; beryllium oxide formed partially in liquid-phase and partially in vapor-phase ... vapor diffuse away to form beryllium oxide spheres that condense on surface resulting in a sphere approximately the same size as the initial specimen.²⁰
 - Heated plates (8-cm × 1.2-cm × 0.075-cm, volume 0.72-cm³) in temperature range 350–950 °C in oxygen²¹:
 - activation energy for oxidation 50,300 cal/mol;
 - minimal dependence on oxygen partial pressure shows limiting factor for oxidation is diffusion of Be vapors through oxide film;
 - oxide film exerts strong effect on reducing vapor pressure – varies with sq. root of oxide layer thickness ... film 99-μg [depth?] lowers vapor pressure 100-fold; limiting factor to oxidation is diffusion through oxide film;
 - at 825 °C Be reacts with oxygen at ~same rate as Zr at 325 °C, at 900 °C Be reacts less with oxygen than Nichrome V ... >600 °C colored oxide forms.
- The O–O₂ flame system was used to investigate the flame temperatures near H₂–O₂ system in the absence of water vapor:
- Dramatically different behavior than H₂–O₂, the *beryllium never ignited*;
 - Beryllium oxide airborne release was 6–7 orders-of-magnitude lower:
 - @ mp Be (1,284 °C), airborne release 1-μg/cm² min; [release rate 10× proposed for large, coherent metal pieces]
 - @ 2,000 °F (1,093 °C), ~10-μg/cm² min;
 - @ 3,600 °F (1,982 °C), ~the release rate was approximately three orders-of-magnitude greater than the release rate at 2000 °F (1,093 °C) (on the order of mg/cm² min);
 - the rate increased exponentially with temperature.
 - Study by Terem – oxidation Be @ 1000 °C greater than for Ni (powder) and 75% complete in 15-min.
 - Canning materials in advanced gas-cooled nuclear reactors; small samples (two pieces – 4-cm × 1.5-cm × 0.75-mm and 3-cm × 1-cm × 0.75-mm):
 - Temperature range – 500–750 °C;
 - <650 °C oxide layer protective rate decreases with time ... after 300-hr at 750 °C, rate initially decreased then increased indicating oxide layer-break away and subsequent non-protective oxidation (Aylmore, Gregg, and Jepson, 1960)³⁹;
 - Experiments show both rapid diffusion flame and slow surface-reaction when beryllium powder ([d_G] 25–32-μm) is injected into a hot [temperature not defined] flame. It appears reasonable to suppose that small particles in a given powder sample are the ones that burn by vapor-phase mechanism. If this interpretation is correct, two conclusions follow. First, large, more heavily protected particles [no data for the assumption that larger particles are more heavily protected] will have to be heated to a higher temperature to ignite than smaller ones ... Ignition parameters are not sharply defined for beryllium particles. Observations are pertinent. There is no defined ambient temperature necessary for ignition; rather, as the ambient temperature increases, more and more particles ignite [no such observation of this phenomenon has been cited in the literature] ...¹⁹;
 - 0.2% [0.002 fraction] of 0.5-g sections of thin Be rods oxidized after 80-min @ 1,185 °C.²¹
 - Burn time of a [d_G] 35-μm particle in an atmosphere of O₂ is 3–4-ms¹⁵: at the bp of beryllium, oxidation is rapid and forms a porous coat [corroborates characteristics of oxide

depend on temperature during formation] of agglomerates of particles in the respirable size range ... fractional airborne depends on mechanical disruption by external action ... Variation in onset accelerated oxidation and ignition may be due to experimental conditions and size and morphology of specimens ... specimen temperature is a balance of heat from exothermic reaction and external sources and losses that depend on particle mass and morphology and experimental configuration ... temperatures cited are typically air-temperature, not specimen temperature; 30- μm specimens probably produce results most representative of powder piles but may indicate a lower temperature due to more rapid rise and better heat retention; ignition range 140–900 °C. Observation – powder shows localized bright spot on ignition rather than uniform ignition corroborates ignition from one point rather than uniform gas temp.

Heated rods, 3/16th-in. diam. (0.48-cm) \times 1/2-in. (1.3-cm) long, in pure O₂ to 1000–1,500 °C of QMV beryllium (from Brush-Wellman, 1.67% oxide + other trace impurities) ... volume 0.226-cm³, mass 0.418-g, area 2.26 cm²; [cited other experiments covering the temperature range from 350 °C to ~1000 °C]. Findings:

- <mp beryllium [to 1,200 °C] – oxidation rate relatively independent of O₂ partial pressure and little oxidation;
- >mp beryllium [>1,400 °C] – demonstrate an approximately sq. root dependence on O₂ partial pressure;
- increasing O₂ pressure \times 6,500 increases the oxidation rate by \times 64;
- <mp beryllium – lack of dependence on O₂ pressure indicates rate-limiting step is diffusion of the Be-vapor through the oxide layer;
- >mp beryllium: oxide layer is non-protective;
- activation energy 70 \pm 15 kcal/mol.

[Equations for oxidation rate presented for just below and above mp Be] is 1,050 °C to 1,200 °C dm/dt

$dt = 0.0179 \text{ mg/cm}^2 \text{ min}$ and remains at that level at the mp = 1,278 °C; at 1,300° to 1,500 °C, >mp Be $dm/dt = 1.29 \text{ mg/cm}^2\text{-min}$ at mp and O₂ partial pressure 144.4-mm (0.19 \times 760 mmHg).⁸

Investigated reaction products as health hazard:

- >50% sample oxidized [size not specified] @ 1,550 °C, 160–480-mm Hg O₂, and 30 min;
- some became white flocculent agglomerates of 1–10- μm [d_G] primary particles;
- in one experiment, oxide was observed on a thermocouple 6-in. above sample;
- oxidation controlled by vapor-phase oxidation, not O₂ dissociation step; or, surface-reaction between chemisorbed Be and O⁻.

Large, coherent metal, >100-g

Jordan⁸ provided an excellent review of the published literature of the impact of thermal stress on beryllium metal (encased in a protective film of “blue” oxide) and deduced the potential airborne release of beryllium oxide when large, coherent pieces of metal are heated by a liquid-petroleum fire with air-augmentation to enhance its combustion to 1260 °C in 30 minutes ([ARF][RF] 3×10^{-6}).

From the published values for the measured airborne release from two separate studies,^{28–30} a surface-specific value of $2 \times 10^{-3} \text{ g Be/m}^2$ was calculated by Jordan⁸ for large, coherent beryllium metal pieces suspended over a liquid-petroleum fire (Appendix B⁸). Jordan recognized that the temperature of the metal core shielded by the thin layer of “blue”, adherent oxide was a balance between the external heat supplied plus the heat generated by the oxidation-reaction and the heat loss to the ambient atmosphere.

In two experimental studies, the beryllium metal was suspended over the fire that is not typical, but bounding, configuration for inadvertent fires in DOE facilities where fires are typically fueled by solid, combustible materials and the beryllium is anticipated to be immersed in the fuel or heated by radiant heat from the fire.

The surface to volume ratio of the SNAP (Space Nuclear Auxiliary Power) configuration used in these tests was $912.1\text{-cm}^2/521.25\text{-cm}^3 = 1.75$ that most closely matches the surface to volume ratio for a sphere but the surface to volume ratio changes with size and the two items are not comparable. On this basis, it is assumed that the airborne release for other shapes (i.e., chips/powder and turnings/swarfs) will be greater on a surface-specific basis.

The details of the two studies are:

1. Everett and Mills January 1963 (also covered in Boyd. January 1963):
 - beryllium reflector in SNAP project;
 - beryllium metal disks 9-in. (23-cm) diameter, 0.5-in. (1.3-cm) thick (implies 964-g beryllium using the density of beryllium) suspended horizontally inside a fire barrel [chimney] with a kerosene-feed fire in pan below + supplemental O₂ to get high temperatures;
 - fire duration 30-min;
 - 15 Hi-Vol samplers in a downwind arc around source, one sampler at top of barrel feed by a right-angle pipe sampling tube;
 - Oxidation increased with increasing temperature:
 - @ 1,260 °C for a 30-min. fire, weight gain 21.44 g [0.44% oxidized (does not specify basis for value)];
 - @ 871 to 982 °C for 32-min fire, weight gain 0.24-g [by analogy to previous value, ~0.005%];
 - only for those tests that yielded disk temperature of 1,093° to 1,260 °C or 30-min fire was airborne contamination detected;
 - sampler on top of barrel yielded 0.05- μg (beryllium?) [inlet duct configuration, sampler-inlet configuration and sampler and chimney flow rate not given, therefore cannot estimate what total emissions may have been] for former and 0.085- μg for the latter [cannot determine which former and latter referred to];
 - swipes from inside the barrel after latter test [cannot determine latter referred to] yielded 0.2- and 1.2- $\mu\text{g}/\text{ft}^2$;

- estimates based on vapor pressure gave higher results;
- conjectured protective oxide layer inhibits vaporization of metal [inhibits release of vapors];

Note: mp Be 1,278 °C that is higher than the disk temperatures achieved during these tests.

Analysis of tests⁸ described in Everett and Mills,³⁰ above:

- beryllium disk 9-in diameter × 0.5-in. thick (22.86-cm diameter × 1.27-cm thick) suspended in a petroleum fire of 30-min duration, metal temperature 1,290 °C [melting point Be 1,284 °C];
- volume = $\pi/4[(22.86\text{-cm})^2][1.27\text{-cm}] = 521.25\text{-cm}^3$;
- mass = [volume][density, 1.85 g/cm³] = $[521.25\text{-cm}^3][1.85\text{-g/cm}^3] = 964\text{-g}$;
- surface area = $[2][\pi/4][(22.86\text{-cm})^2 + \pi[22.86\text{-cm} \times 1.27\text{-cm}]] = 912.1\text{-cm}^2$ or 0.09121-m^2 ; surface: volume = $912.1\text{-cm}^2/521.25\text{-cm}^3 = 1.75$;
- heavy outer oxide layer was powdery but sub-layer next to metal was hard and tightly adherent ... weight gain uncertain due to potential losses during handling but estimate 2% of material oxidized;
- run #4 released most beryllium airborne ... chimney concentration $0.085\text{-}\mu\text{g/m}^3$, only single chimney sampler ... swipes from barrel (chimney) interior yielded μg quantities ... airflow through chimney not given, Jordan estimate [basis unknown] 10-mph ($\sim 5\text{-m/s}$) but could have been much higher (variation in velocity across chimney passing the heated air may be substantial);
- based on 5-m/s air velocity estimate, total beryllium release: diameter of chimney (sacked barrels) 22-in = 55.88-cm, height 62-in. ... X-sectional area = $\pi/4[55.88]^2 = 2,452\text{-cm}^2$ or 0.245-m^2 ;
- volumetric flow = $[0.245\text{-m}^2][5\text{-m/s}] = 1.23\text{-m}^3/\text{s}$;
- for a 30-min sampling, total beryllium release = $[0.85 \times 10^{-6}\text{-g/m}^3][1.23\text{-m}^3][1800\text{-s}] = 1.88 \times 10^{-4}\text{-g}$;

- [material released but deposited in chimney not included];
- release/surface area = $[1.88 \times 10^{-4}\text{-g}]/[0.09121\text{-m}^2] = 2 \times 10^{-3}\text{-g/m}^2$;
- uncertainties:
 - samples of beryllium airborne in chimney from a single location;
 - flow rate is an estimate, not measured;
 - deposition on chimney interior maybe significant but insufficient information to estimate;
 - estimate of release may be an order-of-magnitude [or more] greater.

2. Stewart²⁸ 1961:

- bulk beryllium and plutonium metal specimens were exposed to gasoline fire at 1,080 °C for 30-min;
- the beryllium was in the form of plates, 6-in. × 4-in. × 0.09-in. (15.24-cm × 10.16-cm × 0.2286-cm) and a 200-g rod of plutonium;
- 20 plates of beryllium suspended vertically in chimney above petroleum fire in pan;
- the beryllium plates did not ignite ... the plates warped but retained shape; looked “flaky” and easily broken;
- surface area – $2[15.24\text{-cm} \times 10.16\text{-cm}] + 2[15.24\text{-cm} + 10.16\text{-cm}][0.2286\text{-cm}] = 321.3\text{-cm}^2$;
- for 20 plates, total surface area = $20[321.3\text{-cm}^2] = 6,426\text{-cm}^2 = 0.6426\text{-m}^2$; and,
- volume (20 plates) – $20[15.24\text{-cm}][10.16\text{-cm}][0.2286\text{-cm}] = 707.9\text{-cm}^3$;
- total mass = $[707.9\text{-cm}^3][1.85\text{-g/cm}^3] = 1,310\text{-g}$;
- Estimated mass of the beryllium released was less than plutonium; based on comparison of plutonium and beryllium collected from another test run where both metals were involved ... Jordan assumes equal masses were released; based on the release value in DOE-HDBK-3010-94 of ARF 5×10^{-4} for plutonium from self-sustained oxidation, the release from a 200-g of plutonium is $[200\text{-g}][5 \times 10^{-4}] = 0.1\text{-g}$; (RF = 0.5).
- If 0.1-g of beryllium were released from the 1,310-g Be, ARF = $0.1\text{-g}/$

$1,310\text{-g} = 7.6 \times 10^{-5}$ and surface-specific release = $0.1\text{-g}/0.6426\text{-m}^2 = 0.16\text{-g/m}^2 \approx 0.2\text{-g/m}^2$.

- The mass of beryllium is 7.5× the mass of plutonium but observed that probably less beryllium was deposited (equivalently, airborne) than plutonium (estimate $0.1\text{ [ARF][RF]}_{\text{Pu}}$, $\sim 2.5 \times 10^{-5}$).

Chips/powder, spherical/cubic shape, $d_G < 1.27\text{-cm}$, weight $< 2.0\text{-g}$, surface to volume ratio > 5

There are no specific experimental observations on the oxidation of this generic Be form reported in the literature. The surface to volume ratio of the three generic assumed for Be forms are approximately spheres 6, chips and powders 12, and turnings 82; but, the ratio is somewhat dependent on the size of the form and the specific configuration (the forms assumed are generic and the specific item may deviate from these surface to volume ratios). However, based on technical and engineering judgment and observations of other forms of beryllium material that the oxidation of Be is significantly affected by the surface to volume ratio, certain conclusions such as surface to volume ratio > 5 and ARF/RF values for various accident scenarios are calculated (see Section ‘Chips, Powder [Spherical/Cubic Shape, $d_G < 1.27\text{-cm}$, Weight $< 2.0\text{-g}$, Surface to Volume Ratio > 5]). Investigations of ignition of small pieces (half gram) of Be rods agrees with Macek’s vapor phase condensation & resultant oxide products – rods & droplets.⁸

Turnings, “ribbon-like” shape, weight $< 1.5\text{-g}$, surface to volume ratio > 20

Lindsay and Robinson²⁶: Machining turnings (swarfs) and powder oxidize slowly @ 1,000 °C. At 1,100 °C, oxidizes slowly to nearly standstill as oxide layer thickens ... loses protection as metal melts (1,278 °C) and oxidation rapid.

Blumenthal and Santy¹⁵: Heated 3/16th-in. diam. (0.48-cm) × 1/2-in. (1.3-cm) long rod in pure O₂ to 1000–1,500 °C of QMV beryllium (from Brush-Wellman, 1.67% beryllium oxide + other trace impurities) ... volume 0.226-cm^3 , mass 0.418-g, area 2.26-cm^2 . They cited other experi-

ments covering the temperature range from 350 °C to ~1000 °C. Findings are:

- <mp Be [to 1,200 °C]: oxidation rate relatively independent of O₂ partial pressure and little oxidation;
- >mp Be [>1,400 °C]: sq. root dependence on O₂ partial pressure;
- increasing O₂ pressure × 6,500 increase the oxidation rate by ×64;
- <mp Be: lack of dependence on O₂ pressure indicates rate-limiting step is diffusion of the Be-vapor through the oxide layer;
- >mp Be: oxidation is non-protective;
- Activation energy 70 ± 15 kcal/mol.

Oxidation controlled by:

- vapor-phase oxidation controlled by oxidation, not O₂ dissociation step; or,
- surface-reaction between chemisorbed Be and O⁻.

Nuclear-grade [not defined] beryllium rod 0.5-in. × 3/16-in. (1.3-cm × 0.5-cm), temperature 900–1,500 °C, O₂ pressure 0.5–700-mmHg¹⁵:

- <1,050 °C – Be-O₂ reaction rate slow, decreasing with increasing oxide layer and relatively independent of O₂ pressure;
- between 1,050 and 1,280 °C – reaction rate constant in time and nearly proportional to O₂ pressure (no longer oxide layer-diffusion controlled);
- >mp Be (1,284 °C) rapid oxidation with sq. root dependence on O₂ pressure (O₂ dissociation dependent); and,
- 1,500 °C and 760-mm Hg O₂–50% oxidized within 15-min;
- oxidation rate exponential with temperature in temperature range investigated;
- apparent activation energy <mp Be 60,000 cal/mol (60 kcal/mol); >mp beryllium 70,000 cal/mol (70 kcal/mol);
- no indication of ignition or rapidly accelerating reaction;
- >mp Be sample encrusted with thick, white, porous layer of oxide (the metal core was coated with a thin, dark blue film, pacification

layer) ... porous oxide layer agglomerates of small crystalline platelets a few hundred Angstroms to several μm thick that adhered “fairly tightly” to metal core.

- >mp Be the oxide layer was more flocculent and less adherent ... “Reaction products were found to at a considerable distance from the Be sample”.

Purging the equipment after several experiments, no deposit on filter indicating, perhaps, that the oxide deposit is not readily re-suspended.

Other gases investigated were H₂O-vapor, N₂, nitric oxide, H₂, CO, and CO₂ ... reaction rate water-vapor 2× that of O₂ ... water vapor as a reaction product of fire may be a significant contributor to oxidation of beryllium.

The experiments described in Section ‘Oxidation – experimental observations (literature review)’ on burning beryllium in H₂ + O₂ flame capable of reaching 5,000 °F (2,760 °C) temperature and in the CH₄-air flame are also applicable to turnings/swarfs. For O₂:H₂ 0.33, as the sample reached the melting point of beryllium, significant release of airborne beryllium occurred at the hotspots that were as much as 1,000 °F (538 °C) above bulk temperature of the sample, which reached 3,600 °F (1,982 °C). “In general ignition and subsequent vapor-phase combustion of the type observed with Mg and Al did not occur in the experiments which were run in a fuel-rich flame. The reaction zone appeared to be at, or very near, the surface of the sample, and it seemed likely that the reaction was taking place within the loose porous oxide crust which built up on the surface of the beryllium.”

For O₂:H₂ = 0.99, “... the beryllium samples were found to ignite and burn with a very hot vapor-phase diffusion flame under all but the lowest initial heat flux condition. Samples were ignited under conditions in which the thermal balance, excluding the energy generated by the chemical reaction, would leave the sample at a steady-state temperature below the melting point of the beryllium.”

Transition from reaction near or at the surface to a vapor-phase diffusion flame appeared to take place at a tem-

perature above 3,600 °F (1,982 °C) ... when ignition and vapor-phase oxidation occurred, “... a significant fraction of the total beryllium sample was converted to airborne oxide smoke in a matter of a few seconds.” ... Smoke particle size [*d_G*] up to a few micrometers. Transition from rapid surface-reaction to extremely fast vapor-phase reaction took <0.01-s [10-ms].

Oxidation Conclusions

Conclusions with respect to oxidation experiments presented above are as follows:

- @ temperature <mp Be, water vapor appears to be the most reactive combustion gas;
- @ temperature >2,000 °F (1,093 °C), reaction with O₂ also important;
- *it is not felt, however, that oxygen should be a major reactant with beryllium in an air-supported hydrocarbon fire because the concentration of uncombined oxygen in the flame environment should be low and the beryllium-oxygen reaction is strongly dependent on the oxygen concentration.*

In a typical low-temperature, long-duration, air-supported hydrocarbon fire (water concentration ca. 15%), it appears, in general, that there should be little release of airborne contamination resulting from the long-term exposure (period of minutes) of beryllium components to the fire environment at temperatures up to 2,000 °F (1,093 °C). At 2,000 °F (1,093 °C), however, there will be considerable interaction between the flame environment and the beryllium components, resulting in an oxide reaction product which tends to adhere fairly strongly to the beryllium metal.

Based on the experimental observations it seems unlikely that this reaction product could become dispersed in the air under most conditions, but, it must be kept in mind that the oxide product is made up of agglomerations of airborne-sized particles and it is conceivable that under extreme conditions (a large-scale explosion following a long-duration fire for example) that a

major portion of this reaction product could become airborne.

The degree of interaction between the flame environment and the beryllium components and the amount of airborne contamination that are released will rapidly increase with increasing beryllium metal temperature, concentration in the flame environment of water-vapor and/or oxygen and the total pressure of the environment. In flame environments containing in excess of 20% water vapor at a total pressure of one atmosphere or above, significant amounts of airborne contamination can be expected when beryllium components are heated to or above the melting point of the metal [1,284 °C].

IGNITION ASSUMPTIONS

This purpose of this section is to summarize the experimental data related to ignition of beryllium metal, i.e., self-sustained oxidation, and to select ignition temperatures for recommended ARFs and RFs in Section "Technical basis for airborne release fractions (ARFs)/rates (ARRs) and respirable fractions (RF)". Table 6 lists recorded ignition temperatures for various forms of beryllium metal.

As stated in Section "Oxidation – Experimental Observations (Literature Review)", data and information on the behavior of beryllium metal encased in a thin film of adherent oxide is at times contradictory. Interpretations of ignition temperatures by the authors are as follows:

Large, coherent metal—temperatures reported for the two experiments are "greater than" values; higher value selected >1260 °C. The form of beryllium metal cannot ignite under typical inadvertent fire conditions in non-reactor nuclear facilities. The temperature required to ignite this beryllium form would also affect other materials present such as structural materials and would require extraordinary conditions to heat the metal for a sufficient duration (in the order of hours) for ignition.

Chips/powder—temperatures ranging from 635°C (d_G 37- μ m) to 2,327 °C

(d_G 20–35- μ m particles) have been reported; most value cluster in 700 to 900 °C range; selected temperature of >800 °C as a "conservative" estimate of the ignition temperature. The heat must be applied for a duration of many minutes.

Turnings/swarfs—temperature range for ignition ranged from 780 °C (1–2-mm turnings, other dimensions unspecified) to 1,750 °C [Beryco-10 alloy 125-in. \times 0.25-in. \times 0.015-in. (0.093-cm³)]; a value between the two temperatures extremes is 1000 °C, the ignition temperature selected. The heat must be applied for a duration of minutes.

Dust— d_G , 20- μ m or less. In reality, the material is beryllium metal encased in oxide. In this size range, the oxide constitutes a larger fraction of the weight than for the larger sized beryllium metal forms, above, and some very small sized material may be entirely oxide. The lowest ignition temperature determined, except materials that are coated with a hydrocarbon fuel, is ~750 °C and that value is selected for the ignition temperature of this sized material.

GUIDANCE ON ARF AND RF VALUES APPLICABLE

For emergency planning and process safety management programs, the potential consequences of the release of toxic chemicals have generally focused on volatile compounds. The hazard from the release of chemicals such as beryllium, its oxide, and lithium compounds are particulate materials. As such, the materials released behave as particulate matter and, if their physical and chemical properties are *not* altered by the accident stresses, behave as the surrogate materials used in the experimental studies that are the basis for the DOE-HDBK-3010-94⁵ values. Some aspects of this concept applicable to other chemical particulate releases are discussed by Laul et al.³¹

There are some effects that should be considered such as enhanced release of compounds with lesser densities than the uranium dioxide powder used in the experiments. But the uranium dioxide (and in the cases of the free-fall spill of

powder "slugs" titanium dioxide that has a density of 4.486 g/cm³ versus the density of beryllium oxide that has a density of 3.05 g/cm³) used had a very small particle size distribution that would not typically be found for other materials and somewhat compensates for the minor density effects on the airborne release of particulate material.

There are other properties that may have effects on the airborne release such as the tendency to adhere to other particles in the powder body that are a function of the plasticity of the surface; the size range and morphology of the particle population that may effect the ability to packed and resist de-agglomeration; etc. The effects of the stresses imposed, local conditions, and other parameters, cited above, that determine the airborne release of particulate materials are the interactions of these phenomena and are very complex. The values in DOE-HDBK-3010-94 were generated and evaluated with the intent to provide reasonable bounds for those initiators and materials found in DOE non-reactor nuclear facilities and, with the exceptions cited above, are generally applicable.

Overall, the DOE-HDBK-3010⁵ bounding ARFs and RFs are generally applicable, particularly if physical and chemical properties are not altered by accident stresses such as fires, explosion, spills, loss of confinement, and resuspension. Factors such as density, particle size, and deposition rate play an important role in the evaluation of the ARF/RF values (Laul et al., 2006).³¹

ERPGs/TEELs and Health Effects

Although the ERPG and TEEL values (Table 1) are usually based on total suspended particulate concentrations, credit for a RF in the source term calculation above is also recommended for evaluation of receptors at 100 m or farther from the release point. The RF values should be based on the DOE-HDBK-3010⁵ recommendations (e.g., 0.3 for spill, impact crush, shock vibration, see Table 1) or if known, the source powder distribution (e.g., 0.01 for detonation, deflagration of powder/chips and turnings/swarfs). Larger particles will generally deposit rapidly as the plume travels down wind, such

Table 6. Recorded Ignition Temperatures for Various Forms of Beryllium Metal

Reference	Size	Ignition (°C)
Large, coherent metal		
Stewart ²⁸ (Vixen A)	Plates 200-g	>1080 °C ^a
Everett and Mills ³⁰ , Boyd ²⁹	Disk 964-g	>1260 °C
Chips/powder		
Fairbairn ³⁷	d_G 37- μ m	635–780 °C
	Chips (size not specified)	780 °C
Green ³⁸	Not specified	250 °C ^b
Lindsay and Robinson ²⁶	Chip size not specified	1000 °C
	d_G <37- μ m (<400-mesh)	~600 °C
	d_G 1-mm (1,000- μ m)	~800 °C
	d_G 1,700- μ m	>1,000 °C
Alymore et al. ³⁹	0.4–0.8-g	>700 °C ^c
Reynolds ^{36,40}	0.17-g	945 °C
Gulbransen and Andrews ²¹	0.72-g	950 °C ^c
Blumenthal and Santy ²³	0.5-g	1,264 °C ^c
Macek, Friedman and Semple ²⁵	20–34- μ m	2,327 °C, Ambient gas temp >2,600 °C
Brush-Wellman MSDS	Powder (size not specified)	540°–700 °C
Turnings/Swarfs		
Fairbairn ³⁷	Not specified	1000 °C ^d
Blumenthal and Santy ²³	1–2-mm	780 °C
Kuehl ²⁴	500- μ m wire	~1,000 °C ^e
Green ³⁸	125-in. \times 0.25-in. \times 0.015-in (0.093-cm ³)	1,750 °C ^{f,g}
	Beryco-10 alloy	
Alymore et al. ³⁹	Nuclear-grade [not defined] Be rod 0.5-in. \times 3/16-in. (1.3-cm \times 0.5-cm)	For O ₂ :H ₂ = 0.99, "... the beryllium samples were found to ignite and burn with a very hot vapor-phase diffusion flame under all but the lowest initial heat flux condition. Samples were ignited under conditions in which the thermal balance, excluding the energy generated by the chemical reaction, would leave the sample at a steady-state temperature below the melting point of the beryllium." Transition from reaction near or at the surface to a vapor-phase diffusion flame appeared to take place at a temperature above 3,600° F (1,982 °C) ... when ignition and vapor-phase oxidation occurred.
Be metal dust, <d_G 20-μm		
Lewis and Karlovitz ¹³	d_G 1.21–5.22- μ m	540° d_G 1.21- μ m to 680 °C
Fairbairn ³⁷	Collected on pre-filters	700°–900 °C
	Oil-coated	140° to 280 °C
Lewis and Karlovitz ¹³	1.21–5.22- μ m	Did not by electrical spark
	Thermal atmos.	1.21- μ m @ 910 °C
Green ³⁸	Ave. d_G 1.2- μ m	Cloud 910 °C ^h
		Layer 540 °C ^h
Coleman ¹⁷	2–3-g (size not specified) ^h	
Lewis and Karlovitz ¹³	d_G 1.21- μ m	910 °C

^a Large plates did not ignite.^b Data probably taken from Lewis and Karlovitz.¹³ Also see Jacobson, Cooper, and Nagy.¹⁸^c Accelerated oxidation.^d Some samples had ignition temperatures as low as 140 to 280 °C.^e With ignition, sample temperature rose to >1,000 °C.^f Atypical definition of ignition – temperature where heat input (supplied external heat + heat of reaction) balances heat loss.^g Composition of alloy not specified.^h Thermal energy via furnace; material did not ignite by electrical spark.

that their contribution to an inhalation intake should not be significant to the worker and public. Also, particle sizes larger than 10 μm AED (aerodynamic equivalent diameter) do not penetrate to the deeper regions of the lung and thus are flushed out by the biological removal mechanisms.

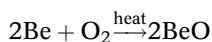
Health effects including exposure limits and regulatory requirements are discussed in detail by Wambach and Laul.³ Only pertinent key points are mentioned briefly here. Particles less than 10 μm AED make it through the respiratory system into the deep lungs, where the fine beryllium aerosol (<10 μm AED) can cause health effects or diseases: (a) acute exposure from ERPG-3, -2 levels, which can be reversible and (b) chronic health effects such as sensitization and chronic beryllium disease, which is caused by mainly due to the immune system of an individual worker. Skin penetration can also cause sensitization and CBD. Thus, not every one who is exposed to beryllium gets sensitized and there is a lag time between the exposure and sensitization in those who get sensitized. Sensitization can occur on a short time scale (e.g., weeks to months) to years after exposure and is precursor to the CBD. The immune system response that results in CBD is generally localized in the part of the lung where respiration occurs and where only particles <10 μm AED deposit. The number of sensitization and CBD cases at each DOE site range mostly from 0 to 4%. The CBD cases are generally far fewer than the sensitization cases. This is because the CBD occurs in fraction of those sensitized. The CBD symptoms may not appear sometime for periods longer than 10 years. The CBD is treatable but not curable.

Threshold limit value (TLV) is $2.0 \mu\text{g}/\text{m}^3$, based on 8-hr time weighted average (TWA) exposure. However, the 10 CFR 850 Rule,³⁵ "Chronic Beryllium Disease Prevention Program; Final Rule", December 8, 1999, requires in Section 850.23 *an action level* at $0.2 \mu\text{g}/\text{m}^3$ (air, 8 hr TWA exposure) for a worker as measured in the worker's breathing zone by personal monitoring," to prevent or reduce the CBD. To protect the public

from CBD, EPA has set a beryllium ambient air limit of $0.01 \mu\text{g}/\text{m}^3$ as a 30-day TWA.³ Thus, health effects consideration is very important working in a beryllium facility (High, Moderate, and Low hazard) to protect the workers and public.

OXIDATION ASSUMPTIONS FOR BERYLLIUM FORMS

As discussed in Section Beryllium oxide, the Be metal particles have very thin coating film (1–8%), called "blue oxide" film that is a protective layer (vapor-diffusion barrier), non-porous and tightly adherent (see Figure 1). Thermal expansion coefficient of Be metal is $11.4 \text{ ppm}/^\circ\text{C}$ and that of BeO is $9.0 \text{ ppm}/^\circ\text{C}$, which causes cracking of the "blue oxide" film on heating. Because of intrinsic expansion in the lattice, at elevated temperatures (>800 $^\circ\text{C}$), vapors diffuse the protective layer and forms different physical characteristics of white "fluffy" material and leads to oxidation:



Oxidation mechanism of beryllium surfaces and films is explained by Adams and Hurd.³² In a fire scenario, as the temperature rises the oxide layer disrupts and the oxidation proceeds rapidly. In addition, fire induced convection current may sweep up the fine metallic powder. However, this fine powder is also likely to get converted to oxide in a fire. *The oxidation largely depends on the temperature, duration of the fire, and the amount of material and its form.* The higher the temperature, the higher is the degree of oxidation.

Fire temperature varies but usually lies in the range of 700–900 $^\circ\text{C}$. Oxidation also takes place rapidly during ignition. The bulk powder, due to large surface area, ignites and is fully oxidized in a short time. There is a wide range of fire and ignition temperatures summarized in Table 6, some in the range of 700–900 $^\circ\text{C}$ and higher. In some experimental studies, ignition temperatures are higher and close to the melting point of beryllium. Turnings/swarfs and large coherent metals have higher ignition temperatures than

chips/powder, based on surface area consideration.

Complete oxidation is generally unlikely in explosions due to the very short time involved (for Be atoms to diffuse or break through the BeO film and then form BeO); however, it also depends on the amount of material and its form. A spill would not involve an oxidation. Materials, depending on the size, at elevated temperatures or close to ignition temperature will lead to oxidation. However, oxidation in a fire at 700–900 $^\circ\text{C}$ depends on the *temperature, duration of fire, and the amount of material and its form.* For example, powder and dust are likely to undergo oxidation, while powder/chips may not undergo complete oxidation. Each process should be evaluated on case-by-case basis. For conservatism, Be metal instead of BeO may be used in terms of ERPG/TEEL values for comparison to estimate consequences to the worker and the public.

TECHNICAL BASIS AND SUMMARY FOR AIRBORNE RELEASE FRACTIONS (ARFS)/RATES (ARRS) AND RESPIRABLE FRACTIONS (RF) OF BERYLLIUM METAL FORMS

Based on the preceding discussions, this section provides the recommended ARFs and RFs for beryllium releases under accident conditions. It is organized by the different forms of beryllium, then by accident stress. The data is summarized in Table 7.

Large, Coherent Pieces of Metal [$>100\text{-g}$]

Explosive releases

Explosion, detonation, in contact with Be: Assumes that the large coherent beryllium metal pieces are fragmented as noted by Dahl and Johnson,³³ and by Shinn et al.³⁴ It is also assumed that any conversion to oxide is fine fragments due to the heat generated by the work performed in the elastic-plastic response that fragments the beryllium metal and is included in the RF fraction cited. However, Jordan⁸ conservatively assumes no oxidation. Depending on the energetics, frag-

ments size may vary, and, so does the degree of oxidation. The material released to the ambient atmosphere from this event can be Be metal and BeO and the ERPG/TEEL values for both are applicable.

- ARF 1E-1, RF 0.3 = [ARF][RF] = 3E-2

Explosion, deflagrations: No documented experimental studies are reported. Assumes that the duration of presence of high temperature is not sufficient to result in ignition; thus, no formation of oxide. Smaller pieces in the respirable size range may be deagglomerated by the pressure imposed. The ERPG/TEEL values for the metal are applicable.

- No significant airborne release, [ARF][RF] <1E-6

Explosive release, over-pressurization of container: For large coherent pieces stored under dry air, not significantly dislodged by venting due to weight of material. For large coherent pieces of beryllium stored under different atmospheres (e.g., inert gases such as nitrogen, helium, argon, etc.) or high relative humidity, these cases must be evaluated on a case-by-case basis. The ERPG/TEEL values for the metal are applicable.

- No significant airborne release, [ARF][RF] <1E-6

Fire (solid, combustible, hydrocarbon fire)

Heated Be metal: When subjected to high temperatures, the surface-specific airborne release is as calculated by Jordan.⁸ Material released to the ambient atmosphere from this event is beryllium metal encased in BeO and the ERPG/TEEL values for the oxide rather than the metal is applicable (oxidation may however depend on the quantity, fire temperature and duration of fire).

- [ARF][RF] = 3×10^{-6} (Jordan⁸ analysis)

Ignited Be metal: Postulated ignition temperature >1,264 °C (potentially @ melting point of beryllium metal, 1,278–1,284 °C). Existing experimental data indicates that ignition is not postulated for representative DOE facility accidents – requisite condition for ignition not attainable. When subjected to high temperatures and based on the experimental observation, >60% of the beryllium remains with the original particle and the assumption is that all the airborne material is released as *oxide* in the μm to sub-μm size range. The ERPG/TEEL values for the oxide rather than the metal is applicable.

- [ARF][RF] = 4E-1

Spill

No significant change in form (possibly some minor disturbance of protective, tightly adherent oxide film that is postulated to rapidly heal upon exposure to air). Beryllium metal encased in “blue oxide” is light-weight and very hard. Some oxide could potential be dislodged from the metal core, but the “blue oxide” is very adherent and no significant fraction is judged to be lost. The ERPG/TEEL values for the metal are applicable.

- No significant airborne release, [ARF][RF] = <1E-6

Crush-impact: No significant change in form, unless seismic response results in impact by large structural component such as a roof panel falling a distance of 20-ft or more (possibly some disturbance of protective, tightly adherent oxide film that is postulated to rapidly heal upon exposure to air). Beryllium encased in “blue oxide” is light-weight and very hard. No significant fraction is judged to be dislodged and suspended. The ERPG/TEEL values for the metal are applicable.

- No significant airborne release = [ARF][RF] <1E-6

Shock-vibration: No significant change in form – BeO-protective layer

tightly adherent to metal core; and no significant fraction is judged to be dislodged and suspended. The ERPG/TEEL values for the metal are applicable.

- No significant airborne release = [ARF][RF] <1E-6

Resuspension: No significant change in form – BeO-protective layer tightly adherent to metal core. The ERPG/TEEL values for the metal are applicable.

- No significant airborne release = [ARF][RF] <1E-6

Turnings/Swarfs [“Ribbon-Like” Shape, Weight <1.5-g, Surface to Volume Ratio >20]

Explosive releases

Explosion, detonation, in contact with Be: Do not postulate significant fragmentation unless very high pressure generated and beryllium on hard, unyielding surface. De-agglomeration and suspension of a fraction of the existing particles in the respirable size range may occur [no information on the RF for this category of beryllium]. Postulate relocation of form due to its’ light-weight. Ignition not postulated due to the short duration that high temperatures are present. Based on the size fraction of beryllium powder provided by the manufacturer, respirable fraction (<7 μm) is about 1%, which is the basis of the proposed ARF/RF value.^{4,6} The ERPG/TEEL values for the metal rather than the oxide are applicable.

- [ARF][RF] = [1.0][0.01] = 1E-2

Explosion, deflagrations: Same discussion as above.

- [ARF][RF] = [1.0][0.01] = 1E-2

Explosive venting due to over-pressurization: Same discussion as above.

- [ARF][RF] = [1.0][0.01] = 1E-2

Table 7. Summary of Encased-Beryllium Metal Airborne Release Fractions and Respirable Fractions

Condition	Airborne Release Fractions/Respirable Fraction Values ^a			
	Large, Coherent Items ^[1]	Powder/Chips ^[2]	Turnings/Swarfs ^[3]	Dust Layer ^[4]
Explosion, detonation	1E-1/0.3 ^b	[ARF][RF] 1E-2	[ARF][RF] 1E-2	[ARF][RF] 4E-1 ^[6]
Explosion, deflagration	[ARF][RF] <1E-6	[ARF][RF] 1E-2	[ARF][RF] 1E-2	[ARF][RF] 4E-1 ^[6]
Explosive release ^[5]	[ARF][RF] <1E-6	[ARF][RF] 1E-3	[ARF][RF] 1E-2	1E-1/0.7
Fire, heated	[ARF][RF] 3E-6 ^{[6]c}	[ARF][RF] 1.5E-5 ^{[6]c}	[ARF][RF] 2.4E-4 ^{[6]c}	[ARF][RF] 3E-4 ^[6,10]
Fire, ignited	[7]	[ARF][RF] 4E-1 ^[6]	[ARF][RF] 4E-1 ^[6]	[ARF][RF] 4E-1 ^[6,10]
Fire, packaged combustible waste, waste ignited, Be heated	–	[ARF][RF] 1.5E-5 ^[6,8,10]	–	[ARF][RF] 3E-4 ^[6,9,10]
Fire, packaged combustible waste, waste and Be ignited	–	–	–	[ARF][RF] 4E-1 ^[6,9]
Free-fall spill ^d	[ARF][RF] <1E-6	[ARF][RF] <1E-6	[ARF][RF] <1E-6	2E-3/0.3
Crush-impact ^d	[ARF][RF] <1E-6	[ARF][RF] <1E-6	[ARF][RF] <1E-6	1E-3/0.3
Shock-vibration ^d	[ARF][RF] <1E-6	[ARF][RF] <1E-6	[ARF][RF] <1E-6	1E-3/0.3
Resuspension ^d	[ARF][RF] <1E-6	[ARF][RF] <1E-6	[ARF][RF] <1E-6	ARR 4E-5/hr

[1] Characteristics and ignition temperature for Large, Coherent Metal – weight >100-g; do not ignite under conditions found in typical DOE facility fires (fire temperature and duration of application of heat).

[2] Characteristics and ignition temperature for Powder/Chips – cubic/spherical shape d_G <1.5-cm weighing <1.5-g; surface:volume >5; ignition temperature >800 °C applied over period of minutes.

[3] Characteristics and ignition temperature for Turnings/Swarfs – “ribbon-like: form weighing <1.5-g; surface:volume >20; ignition temperature >1000 °C applied over period of minutes.

[4] Characteristics and ignition temperature for Dust – d_G , 10-μm, ignition temperature 750 °C.

[5] For over-pressure 500-psig and less.

[6] ERPGs/TEELs for oxide apply.

[7] Large, coherent pieces of beryllium metal do not ignite under DOE facility fire conditions. If fire temp exceeds 1264 °C, then ARF/RF value of 4E-1 applies.

[8] Released to free volume of sealed, non-combustible (55-gal metal), drum.

[9] Postulated only if metallic surfaces with fine powder contamination have not been decontaminated and are known to be placed in a waste package.

[10] The ARF and RF for this release calculates the amount of Be metal released. If the material is released in air, the material is converted to oxide. To compare the release to the ERPG/TEEL for oxide, the calculated mass must be increased by a factor of 2.77.

^a See discussion in Section ‘Physical Characteristics’.

^b Depending on the energetics, fragment sizes may vary; both Be metal and BeO may apply.

^c If the material quantity is large, then oxidation may not be complete. ERPG/TEEL of Be metal may apply.

^d ARF/RF values of <1E-6 for spill, impact, vibration and resuspension are extremely low.

Fire (solid, combustible, hydrocarbon fire)

Heated Be metal-BeO: If immersed in burning combustible material, oxygen availability low and formation of oxides retarded until beryllium vapors reach ambient atmosphere. Reactions with combustion products are unknown. Typically, in fires involving solid, combustible materials, heat is transferred by radiation and would only impact a portion of the beryllium-metal/oxide surfaces. The ARF and RF value is increased by the surface to volume ratio between the large, coherent metal and a conservative estimate of increased surface to volume ratio for turnings/swarfs, 80. Material released to the ambient atmosphere from both heating and from the ignited metal is beryllium oxide.

- $[ARF][RF] = 80$
 $[3 \times 10^{-6}] = 2.4E-4$

Ignited encased-beryllium metal: postulated ignition temperature 1,000 °C. Based on the experimental observation that >60% of the beryllium remains with the original particle, the assumption is that all the airborne material is released as *oxide* in the μm to sub- μm size range:

- $[ARF][RF] = [4E-1][1.0] = 4E-1$

Spill

Particles in this category of encased-beryllium metal are too large to be suspended as a consequence of free-fall through air of 10-ft. The characteristics and mass of form is insufficient to result in significant change in form upon impact with hard, unyielding surface (possibly some disturbance of protective, tightly adherent oxide film that is postulated to rapidly heal upon exposure to air). The ERPG/TEEL values for the metal are applicable.

- airborne release, $[ARF][RF] < 1E-6$

Crush-impact: No significant change in form, unless seismic response results in impact by large structural component such as a roof panel falling a distance of 20-ft or more

(possibly some disturbance of protective, tightly adherent oxide film that is postulated to rapidly heal upon exposure to air). The ERPG/TEEL values for the metal are applicable.

- No significant airborne release, $[ARF][RF] < 1E-6$

Shock-vibration: No significant change in form. BeO-protective layer tightly adherent to metal core. The ERPG/TEEL values for the metal are applicable.

- No significant airborne release, $[ARF][RF] < 1E-6$

Resuspension: Material in this category is too large for suspension (can be made airborne but do not remain suspended). No significant change in form. BeO-protective layer is tightly adherent to metal core. The ERPG/TEEL values for the metal are applicable.

- No significant airborne release, $[ARF][RF] < 1E-6$

Chips, Powder [Spherical/Cubic Shape, $d_g < 1.27\text{-cm}$, Weight $< 2.0\text{-g}$, Surface to Volume Ratio > 5]

Explosive releases

Explosion, detonation, in contact with beryllium powder pile: Do not postulate significant fragmentation, unless very high pressure generated and beryllium on hard, unyielding surface. Postulated relocation of form is due to its light-weight. Ignition not postulated due to the short duration that high temperatures are present, but dislodgement of particles is in the respirable size range. Based on the size fraction of beryllium powder provided by the manufacturer, respirable fraction ($< 7 \mu\text{m}$) is about 1%, which is the basis of the proposed ARF/RF value.^{4,6} The ERPG/TEEL values for the metal rather than the oxide are applicable.

- $[ARF][RF] = 1E-2$

Explosion, deflagrations: No pertinent experimental data found. Same discussion as above. The ERPG/TEEL

values for the metal rather than the oxide are applicable.

- $[ARF][RF] = 1E-2$

Explosive Release, Venting due to over-pressurization of container, and Venting due to over-pressurization of container: No pertinent experimental data found. Assume form relocated by pressure generated. Material released to the ambient atmosphere from this event is encased-beryllium metal and, due to the uncertainty of the material released, the ERPG/TEEL values for the metal rather than the oxide are applicable.

- $[ARF][RF] = [1E-1][0.01] = 1E-3$
(DOE-HDBK-3010-94 value for release from venting pressurized powder and RF from source material)

Fire (solid, combustible, hydrocarbon fire [postulated ignition temperature for this beryllium form is $> 800^\circ\text{C}$])

Encased-beryllium metal particle (heated to $< \text{ignition temperature}$): Assume that the beryllium metal is exposed to thermal energy sufficient to accelerate the vapor-diffusion through the "blue oxide" layer but not sufficient to ignite the metal. Assume the airborne release is proportional to the surface to volume ratio between the powder and the large, coherent metal tested, as cited, above. Material released to the ambient atmosphere from both heating and from a small fraction that may be ignited is beryllium oxide.

- $[ARF][RF] = 5 [3E-6] = 1.5E-5$

Encased-beryllium metal particle ignited ($> 800^\circ\text{C}$): Assume that the beryllium metal is exposed to thermal energy sufficient to ignite the metal, $> 800^\circ\text{C}$. The airborne release of oxide is that cited by experimenters for the fraction of the initial weight that remains with the original piece is $> 60\%$. Material released to the ambient atmosphere from both heating and from the ignited metal is beryllium oxide and the ERPG/TEEL values for the oxide rather than the metal are applicable.

- $[ARF][RF] = [4E-1][1.0] = 4E-1$

Encased-beryllium metal -surface-contaminated, combustible wastes [Particle size of beryllium contaminant $>d_G$ 20- μm ($>\text{dust}$)]: It is assumed that the temperature of a solid, combustible material-fueled fire is insufficient to ignite beryllium pieces in this size range. It is further assumed that such pieces will oxidize and release oxide to the container in which the waste is contained. Waste would provide internal heat for oxidation. Materials released to a non-combustible container are assumed to moderate the release of the airborne materials to the ambient atmosphere. Material released to the ambient atmosphere from both heating and from the ignited metal is beryllium oxide and the ERPG/TEEL values for the oxide rather than the metal are applicable.

- $[\text{ARF}][\text{RF}] = 5 \quad [3\text{E}-6] = 1.5\text{E}-5$
(released to contained volume)

Spill

Particle in this category of encased-beryllium metal are too large to be suspended as a consequence of free-fall through air of 10-ft. Mass of form is insufficient to result in significant change in form upon impact with hard, unyielding surface (possibly some disturbance of protective, tightly adherent oxide film that is postulated to rapidly heal upon exposure to air). Material released to the ambient atmosphere from this event is encased-beryllium metal and the ERPG/TEEL values for the metal rather than the oxide are applicable.

- No significant airborne release, $[\text{ARF}][\text{RF}]$ value of $<1\text{E}-6$

Crush-impact: Same discussion as above for crush-impact. The ERPG/TEEL values for the metal are applicable.

- No significant airborne release, $[\text{ARF}][\text{RF}] <1\text{E}-6$

Shock-vibration: No significant change in form; the beryllium oxide-protective layer adheres tightly to the

metal core. The ERPG/TEEL values for the metal are applicable.

- No significant airborne release, $[\text{ARF}][\text{RF}] <1\text{E}-6$

Resuspension: Same discussion as above for resuspension. The ERPG/TEEL values for the metal are applicable.

- No significant airborne release, $[\text{ARF}][\text{RF}] <1\text{E}-6$

Dust, Particles $d_G < 20\text{-}\mu\text{m}$; Dust Layer

Explosive Releases

Explosion, detonation, in contact with beryllium: It is assumed that, due to its size, beryllium metal in this size range is ignited by the heat generated by the detonation. The residual oxide particles remaining are assumed to be approximately the same size as the initial particles. Although the d_G of the beryllium oxide particles is up to 20- μm , due to its density of 3.025 g/cm³, the d_{AED} of the residual particles is $20[\sqrt{3.025}] = \sim 35\text{-}\mu\text{m}$. Therefore, it is assumed that 60% of the oxide formed remains with the original particle and is not released. Material released to the ambient atmosphere from the ignited metal is beryllium oxide and the ERPG/TEEL values for the oxide rather than the metal are applicable.

- $[\text{ARF}][\text{RF}] = [4\text{E}-1][1.0] = 4\text{E}-1$

Explosion, deflagrations: No experimental data found. Same discussion as above. The ERPG/TEEL values for the oxide rather than the metal are applicable.

- $[\text{ARF}][\text{RF}] = [4\text{E}-1][1.0] = 4\text{E}-1$

Explosive release, over-pressurization of container (no heat source involved): It is assumed that this phenomenon does not modify the physical characteristics of beryllium in this size category. Since the surrogate material used in the experimental studies had a MMD of d_G of 1- μm , the pressurized release values cited in DOE-HDBK-

3010-94 are applicable. Material released to the ambient atmosphere from this event is encased-beryllium metal and the ERPG/TEEL values for the metal rather than the oxide are applicable due to no heat source involved.

- $[\text{ARF}][\text{RF}] = [1\text{E}-1][0.7] = 7\text{E}-2$

Fire (solid, combustible, hydrocarbon fire)

Encased-beryllium metal: Encased-beryllium metal particle (heated to $<\text{ignition temperature } >750^\circ\text{C}$) due to smaller particle size: assume that the beryllium metal is exposed to thermal energy sufficient to accelerate the vapor-diffusion through the "blue oxide" layer but not sufficient to ignite the metal. Assume the airborne release is proportional to the surface to volume ratio (no ratio has been established and a value greater than for other forms, 100, is applied) between the dust particles and the large, coherent metal tested, as cited, above. Material released to the ambient atmosphere from both heating and from the ignited metal is beryllium oxide, and the ERPG/TEEL values for the oxide rather than the metal are applicable.

- $[\text{ARF}][\text{RF}] = 100 [3\text{E}-6] = 3\text{E}-4$

Ignited metal: Encased-beryllium metal particle (heated to $>\text{ignition temperature } >750^\circ\text{C}$): Assume that the beryllium metal is exposed to thermal energy sufficient to sufficient to ignite the metal. Assume the airborne release is as cited for other beryllium metal forms, $<40\%$, but, due to the size distribution of the source material, it is assumed that all of the released material is in the respirable size range. Material released to the ambient atmosphere from both heating and from the ignited metal is beryllium oxide and the ERPG/TEEL values for the oxide rather than the metal are applicable.

- $[\text{ARF}][\text{RF}] = [4\text{E}-1][1.0] = 4\text{E}-1$

Be-surface-contaminated, combustible wastes in non-combustible container: Encased-beryllium surface-

contaminated, combustible wastes [particle size of beryllium contaminant is $<d_G$ 20- μm (*dust*)]: Any airborne release of oxide is to the contained volume, unless, the containment fails. It is assumed that the temperature of a solid, combustible material-fueled fire may/may not be sufficient to ignite beryllium pieces in this size range. It is further assumed that ignited pieces will oxidize and release oxide to the container. Materials released to a non-combustible container are assumed to moderate the release of the airborne materials to the ambient atmosphere. Material released to the ambient atmosphere from both heating and from the ignited metal is beryllium oxide and the ERPG/TEEL values for the oxide rather than the metal are applicable.

- Heated beryllium contamination: $[\text{ARF}][\text{RF}] = 3\text{E}-4$
- Ignited beryllium contamination: $[\text{ARF}][\text{RF}] = [4\text{E}-1][1.0] = 4\text{E}-1$

Spill

There is no experimental data known. Beryllium particles in this size range (d_G 20- μm and less) are similar in size distribution to the material used in the experimental basis for airborne release values cited in DOE-HDBK-3010-94 and the values are applicable. No significant change in form. Material released to the ambient atmosphere from this event is encased-beryllium metal and the ERPG/TEEL values for the metal rather than the oxide is applicable.

- $[\text{ARF}][\text{RF}] = [2\text{E}-3][0.3] = 6\text{E}-4$

(The bounding values for 3-m spills of powder in DOE-HDBK-3010-94 does not exceed the RF in the source powder of 0.01).

Crush-impact: No significant change in form, unless seismic response results in impact by large structural component such as a roof panel falling a distance of 20-ft or more. Particles in this size category will be suspended by shock-vibration response of the substrate and the surface turbulence induced by large falling objects. Although much smaller than

the size of powder used in the experimental study cited in DOE-HDBK-3010-94, beryllium metal particles in this size category are postulated similarly to the that cited in the handbook using the maximum measure values for the respirable fraction (rather than 10% RF recommended in DOE-HDBK-3010-94). Material released to the ambient atmosphere from this event is encased-beryllium metal and the ERPG/TEEL values for the metal rather than the oxide is applicable.

- $[\text{ARF}][\text{RF}] = [1\text{E}-3][0.3] = 3\text{E}-4$

Shock-vibration: No significant change in form. The beryllium oxide-protective layer is tightly adherent to the metal core. Particles in this size category will be suspended by shock-vibration response of the substrate and the surface turbulence induced by large falling objects. Forces are generated by shock-vibration (flexing of the substrate). Same discussion as above. The ERPG/TEEL values for the metal rather than the oxide are applicable.

- $[\text{ARF}][\text{RF}] = [1\text{E}-3][0.3] = 3\text{E}-4$

Resuspension: No significant change in form. BeO-protective layer is tightly adherent to metal core. Beryllium metal in this size range will be suspended by the forces and turbulence generated by gases passing parallel over the surface. The ARR values cited in DOE-HDBK-3010-94 are applicable. Material released to the ambient atmosphere is beryllium metal encased in BeO and the ERPG/TEEL values for the metal rather than the oxide are applicable.

- $[\text{ARF}][\text{RF}] = 4\text{E}-5/\text{hr}$

Dust Suspended (Airborne Cloud)

Material in this category is by definition airborne and a substantial fraction is in the respirable size range. Material in this category is d_G 20- μm or less. The conservative assumption is that all the material in this instance is in the respirable size range. The ARF and RF are limited to venting pressures of <500 psig. The ERPG/TEEL values

for the metal rather than the oxide are applicable.

- ARF 1E + 0, RF 1.0

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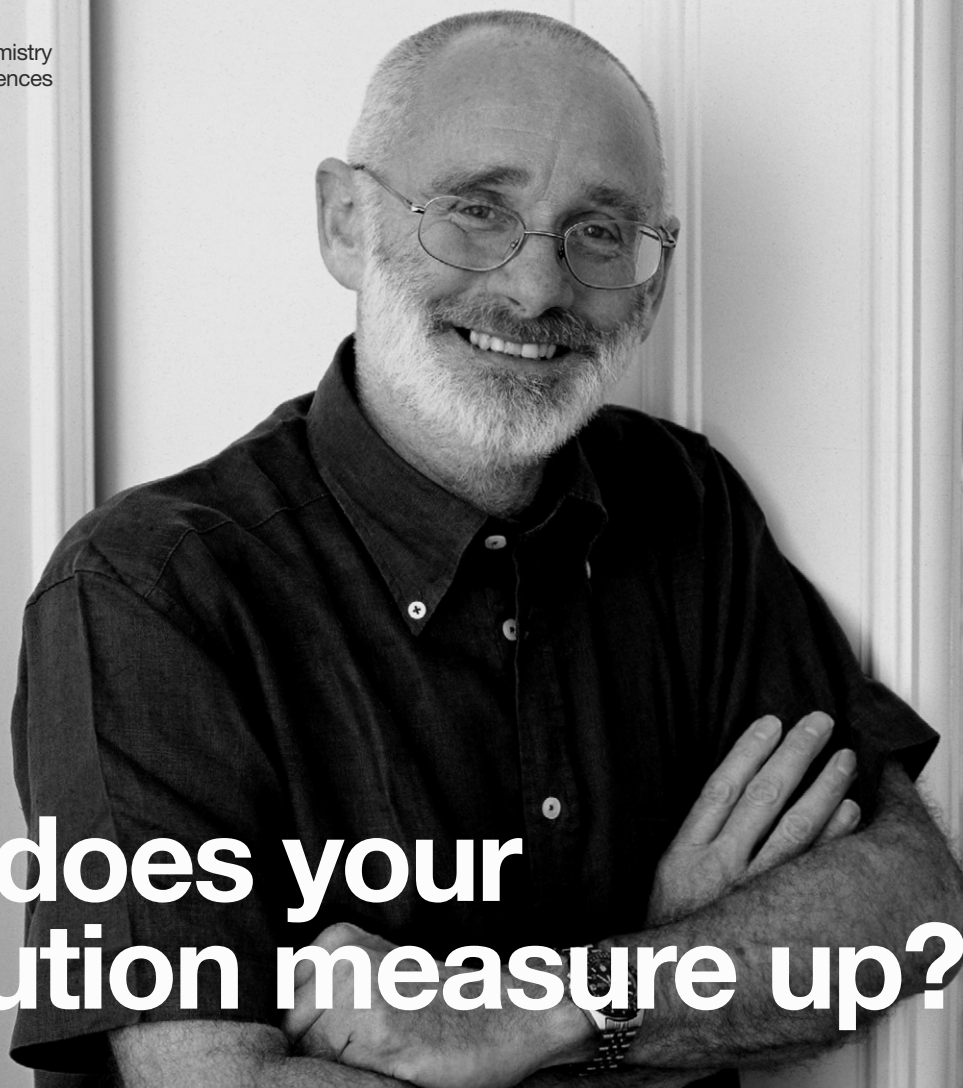
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